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A TWO STAGE RESPIRABLE AEROSOL SAMPLER USING NUCLEPORE FILTERS IN SERIES

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Abstract—Respirable and nonrespirable particles are separately collected from the atmosphere in a size selective sampler consisting of two Nuclepore filters in series. The first filter has pore diameters of $12\text{ }\mu\text{m}$ and collects, to a close approximation, the nonrespirable fraction as defined by the ACGIH criteria. The second filter has pore diameters of $0.2\text{ }\mu\text{m}$ and collects the respirable fraction. Experimental measurements of the efficiency of the $12\text{-}\mu\text{m}$ filter are found to be in good agreement with theoretical predictions based on calculated particle trajectories in a flow field that satisfies the Navier-Stokes equations. Atmospheric samples were collected using both the techniques of series filtration and of virtual impaction. A mass and elemental analysis of the aerosol deposits using gravimetric and X-ray fluorescence techniques demonstrated that the results from the two types of sampler are comparable. Filter loading as great as $90\text{ }\mu\text{g}/\text{cm}^2$ was investigated and found to produce no change in filter performance as measured by the location of the 50% cutpoint.

INTRODUCTION

To evaluate the health aspects of particles suspended in the atmosphere, the mass and composition of the particles present in various size ranges must be determined. Because the cost of the analysis increases as the number of size ranges increases, it is prudent to fractionate the particles into only the required number of size ranges. In 1952, the British Medical Research Council adopted a definition of respirable dust in terms of percent respirable vs diameter for spheres of unit density, and in 1968, similar criteria were established by the American Conference of Governmental Industrial Hygienists (ACGIH), (Aerosol Technology Committee, 1970; Lippman, 1970). These criteria are extended to particles of other than unit density by replacing geometric diameter with aerodynamic diameter, which is defined to be the product of diameter and the square root of specific gravity. Particles of other than spherical configuration may be categorized in terms of an equivalent aerodynamic diameter calculated for a spherical particle exhibiting similar aerodynamic characteristics. This permits atmospheric particles to be fractionated into two size ranges, respirable and nonrespirable. The ACGIH criteria include specifications of collection efficiency vs aerodynamic diameter for a size selective sampler of airborne respirable dust.

Lippman (1970) has reviewed a variety of techniques for the sampling of respirable particles. One is the use of a cyclone separator followed by a backup filter (Lippman and Harris, 1962). The cyclone collects the nonrespirable fraction with an efficiency that closely matches the ACGIH criteria, allowing the backup filter to collect the respirable fraction. Roesler (1966) has explored the use of a polyurethane foam prefilter followed by a backup filter as a size selective device. By proper choice of foam thickness and porosity and of air flow rate, the particle fractionation can be made to closely approximate the ACGIH criteria but the device suffers from the fact that the fractionation changes as the foam prefilter becomes laden with particles.

Unfortunately, it is difficult to make gravimetric or chemical measurements of the respirable fraction collected in either the cyclone or foam filter. A size selective sampler which operates on the principle of virtual impaction and lends itself to both chemical and gravimetric analysis has been described by Dzubay and Stevens (1975). The device is designed to match the ACGIH criteria at the 50% cutpoint, but does not exactly match the criteria at other points on the collection efficiency curve.

The work described here is an effort to develop a fractionating sampler that consists of two Nuclepore filters (Nuclepore Corp., Pleasanton, Calif.) in series. The first filter has pores of large diameter to collect the nonrespirable fraction; the second has pores of $0.2\text{ }\mu\text{m}$ to collect the respirable fraction. The Nuclepore filters consist of $10\text{-}\mu\text{m}$ thick sheets of polycarbonate through which cylindrical pores have

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† Mention of commercial products does not constitute endorsement by the U.S. Environmental Protection Agency.

been etched (Spurny *et al.*, 1969a). The filters have high chemical purity and good mass stability making them well suited for chemical and gravimetric analysis of the collected particles. Since the particle fractionation takes place at the filter surface, this technique is particularly well suited to miniaturization. Although Spurny *et al.* (1974) have investigated the problem of clogging for a Nuclepore filter and although their results might point to the fractionation process described here becoming a function of filter loading, our preliminary investigations do not reflect any such dependency within the constraints of 24 to 48 hour atmospheric sampling or a maximum deposition of $90 \mu\text{g}/\text{cm}^2$.

The idea of using a sequence of Nuclepore filters to determine size distribution was originally proposed by Spurny *et al.* (1969b). Melo and Phillips (1975) have recently described a procedure for determining the size distribution of aerosols by means of several Nuclepore filters of varying pore sizes in series. In the present work only two filters are used in series, and the primary effort is devoted to matching the collection efficiency of a Nuclepore filter to the ACGIH collection efficiency criteria for a size selective sampler. This required filters with pore diameters of $12 \mu\text{m}$. Such filters were developed for the project by Nuclepore Corporation. The present work does not deal with the question of pressure drop across the filter surface, however, this question has been treated recently by Liu and Kuhlmeier (1976).

THEORETICAL

Spurny *et al.* (1969b) have modeled the Nuclepore filter for the collection of submicron particles. They considered the filter to be a bundle of parallel, equally spaced, uniform diameter circular capillaries, oriented perpendicular to the filter surface. Particle deposition was considered to be the result of three mechanisms:

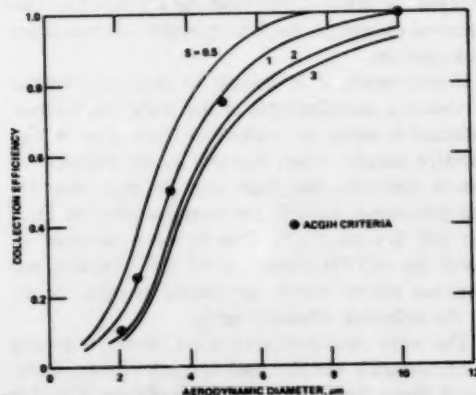


Fig. 1. Theoretical collection efficiency vs. aerodynamic diameter for a $12 \mu\text{m}$ pore diameter Nuclepore filter (10% porosity) presented with respect to particles of various specific gravities, sampled at a face velocity of 12.7 cm/sec . The ACGIH criteria for sampling respirable particles are indicated.

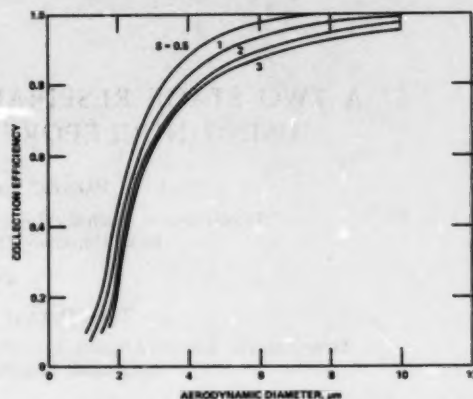


Fig. 2. Theoretical collection efficiency vs. aerodynamic diameter for a $12 \mu\text{m}$ pore diameter Nuclepore filter (10% porosity) presented with respect to particles of various specific gravities, sampled at a face velocity of 38.1 cm/sec .

inertial impaction, interception, and diffusion. The partial efficiency attributable to impaction was determined from the formula derived by Pich (1964), which assumed a parabolic flow field. Smutek and Pich (1974) and Smith and Phillips (1975) have discussed the inaccuracies and limitations of this assumed flow field. In the present study, the same description of the filter has been assumed, but a more accurate flow field has been used. The flow field was obtained from a numerical solution to the Navier-Stokes equations for laminar flow through a cylindrical pore, following a method similar to that used by Vrentas and Duda (1973). Trajectories of particles in this flow field were obtained numerically in a manner similar to that used by Marple and Liu (1974). From the trajectories, the collection efficiency curves were determined; the assumption was made that a particle is collected whenever its center of mass passes within a particle radius of the filter surface. Only particles with diameters greater than $1 \mu\text{m}$ were considered in order to exclude diffusion and noncontinuum effects. The above theory is presented in detail by Parker (1975).

Efficiency curves calculated from this theory are plotted as a function of aerodynamic particle diameter in Figs. 1 and 2 for a Nuclepore filter with a $12 \mu\text{m}$ pore diameter and a porosity of 10%. Efficiency curves are shown for values of specific gravity, $S = 0.5, 1, 2$, and 3 . The curves in Fig. 1 are calculated for a face velocity of 12.7 cm/sec . The face velocity is defined to be the volumetric air flow rate divided by the exposed area of the filter. The circles in Fig. 1 represent the ACGIH criteria for sampling nonrespirable particles. There is excellent agreement between the calculated efficiencies and the ACGIH criteria for particles with a specific gravity of unity. The agreement is particularly good near the 50% cut-point. However, there is some deviation for other specific gravities. If particle deposition were determined only by inertial impaction, as in an ordinary impactor where the characteristic dimension of the impactor

is much greater than the particle diameter, then the curves would have been independent of specific gravity. However, the interception of particles by the small pores is an important collection mechanism for the filter, and this causes the curves to differ.

The curves in Fig. 2 are calculated for a face velocity of 38.1 cm/sec. At this flow rate, the 50% cutpoint occurs at a diameter of 2.3 μm for a particle having a specific gravity of 1.0. The spread in the curves for different specific gravities is about half as wide in Fig. 2 as it is in Fig. 1. As the increased flow rate shifts the 50% cutpoint toward smaller diameters, the effect of interception on the collection efficiency is noticeably smaller.

It is necessary to consider the possibility that particles could bounce after impaction and pass through the pores of the filter. The probability of bounce depends upon particle size and flow rate. Dahneke (1973) has reported experimental data on particle bounce for uncharged, 1.27- μm diameter polystyrene latex spheres striking the target at normal incidence. He found the capture limit velocity (the velocity above which bouncing predominated and below which sticking usually occurred) to be 83 and 91 cm/sec, respectively, for polished stainless steel and polished quartz surfaces. Table 1 lists typical impact velocities and impact angles for theoretical particles impacting on a 12- μm pore diameter Nuclepore filter (10% porosity). These figures were taken from the computer solutions for particle trajectories. In most cases the impact velocities are well below the capture limit velocities determined by Dahneke. However, the impact angles are relatively large, and particles could be bouncing at lower velocities than would apply at normal incidence. Because the impact velocities range from only 18 to 34 cm/sec for a face velocity of 12.7 cm/sec, it may be expected that the probability of bounce is small for this flow rate. However, more quantitative information concerning the bouncing of particles on a Nuclepore surface is needed, especially if the filters are used at higher flow rates where the impact velocities are close to or exceed 80 cm/sec.

EXPERIMENTAL

Collection efficiencies of the 12- μm pore diameter filter were measured using monodisperse aerosols of fluorescent dye. The aerosols were produced by means of a vibrating orifice generator (Berglund and Liu, 1973) equipped with a 10- μm orifice. In this generator, a stream of rhodamine B fluorescent dye and dioctyl phthalate oil (in equal proportions by volume) dissolved in methanol was pumped through the orifice to form droplets. The droplets were dispersed in dry air to prevent agglomeration and to evaporate the methanol. The actual size of the particles was calculated from the frequency of vibration and the concentration of dye and oil in the methanol (Berglund and Liu, 1973). The generated aerosol was passed through a cylindrical ^{85}Kr radiation source to reduce the electrostatic charge on the particles. Particles were collected from this aerosol using 12 and 0.4 μm pore diameter Nuclepore filters in series. The dye was extracted from the filters using methanol and analyzed using a fluorimeter. The filter efficiency was determined from the amount of fluorescent dye collected on each filter.

Atmospheric aerosols were collected outdoors on the roof of a building in downtown Durham, North Carolina, population ca. 100,000. In addition to the Nuclepore filters in series, a virtual impaction type of dichotomous sampler fabricated by Environmental Research Corporation was operated for comparison. The virtual impactor had a 50% cutpoint at 3.5 μm and used Fluoropore filters to collect the fine and coarse particle fractions. Flow rates were controlled with critical flow orifices for the filters in series and with a servomechanism for the virtual impactor. Flow rates were measured at the beginning and end of each run by means of either a dry test meter or an anemometer.

RESULTS AND DISCUSSION

The measured filtration efficiencies for a laboratory-generated aerosol are compared with theoretical predictions in Fig. 3. There is excellent agreement for particles smaller than 5 μm in diameter. For larger particles, the measured efficiencies are lower than the calculated values by about 10%. Such deviations are in excess of the estimated 3% accuracy of the measurements. A possible cause of the deviations is that the larger particles may bounce when they impact on the filter surface, causing reentrainment and subsequent penetration through the filter.

Table 1. Theoretical impact velocities and impact angles obtained from the numerical solutions for particle trajectories

Face velocity (cm/sec)	Particle diameter (μm)	Particle specific gravity	Impact velocity (cm/sec)	Impact angle (from normal) (deg)
12.7	1	1	34	49.2
	3	1	34	41.3
	10	1	28	3.5
	1	4	22	54.1
	3	4	27	26.6
	10	4	18	1.6
25.4	1	1	56	35.3
	3	1	61	53.1
	10	1	43	2.5
38.1	1	1	75	55.8
	3	1	85	30.3
	10	1	57	1.9

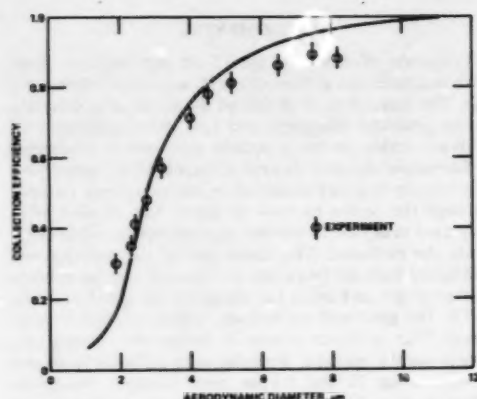


Fig. 3. Experimental and theoretical collection efficiency vs. aerodynamic diameter for a 12 μm pore diameter Nuclepore filter (10% porosity) presented with respect to particles of specific gravity 1.1, sampled at a face velocity of 25.4 cm/sec.

The effect of particle loading on the filter efficiency was also investigated. Three 12- μm pore diameter Nuclepore filters were used outdoors to collect atmospheric particles for periods of 24 to 48 hours at flow rates of 6 to 10 l/min, and the mass loadings were determined gravimetrically. After collecting the

atmospheric particles, these filters were used to collect a laboratory generated rhodamine B aerosol, and collection efficiencies for the rhodamine aerosol were determined. For comparison, the rhodamine B aerosol was collected with each of two clean filters and the collection efficiencies determined. The results of these measurements are listed in Table 2, and they indicate that mass loadings as high as 90 $\mu\text{g}/\text{cm}^2$ do not effect the collection efficiency. For a sampling period of 24 hours, this maximum loading is equivalent to a 100- $\mu\text{g}/\text{m}^3$ concentration of coarse particles in the atmosphere (assuming a sampling face velocity of 12.7 cm/sec). Further measurements are needed at higher loadings to determine the point at which the efficiency of the filter becomes affected.

The results of the field test of the series filtration technique are presented in Table 3. From the elemental analysis, it appears that most of the Si, Ca, Ti, and Fe occur as large particles, while most of the S, Zn, Br, and Pb occur as small particles. This result is consistent with the concept of a bimodal distribution of atmospheric particles with coarse particles consisting of mechanically generated soil dust and fine particles consisting of combustion and secondary aerosols (Whitby *et al.*, 1972). Table 3 also shows fairly good agreement for the fine and coarse fraction-

Table 2. The effect of particulate mass loading on the collection efficiency of a 12 μm pore diameter Nuclepore filter with 10% porosity

Filter	Loading time (h)	Loading flow rate (l/min)	Particulate mass collected (μg)	Particulate loading ($\mu\text{g}/\text{cm}^2$)	Collection efficiency* (%)
1	0	0	0	0	56
2	0	0	0	0	54
3	24	6	110	13	56
4	24	6	150	19	58
5	48	10	730	91	56

* The collection efficiencies were measured at a face velocity of 12.7 cm/sec for 3.5- μm diameter particles having a specific gravity of 1.1.

Table 3. Comparison of mass and elemental composition for aerosols collected in the series filtration and virtual impaction types of dichotomous samplers operated in Durham, North Carolina, for the 24 hr period beginning at noon, January 20, 1976

Element	Total* ($\mu\text{g}/\text{m}^3$)		Fine fraction (%)	
	Filters in series†	Virtual impactor	Filters in series‡	Virtual impactor
Si	4.1	5.1	12	9
S	2.7	2.5	74	83
K	0.51	0.62	29	28
Ca	0.72	0.95	7	11
V	0.09	0.10	66	71
Fe	1.0	1.4	13	17
Zn	0.09	0.09	52	65
Br	0.54	0.55	68	74
Pb	1.8	1.8	68	74
mass†	48	64	51	44

* Sum of fine and coarse particle concentrations.

† Total mass determined gravimetrically.

‡ Sampled at a face velocity of 12.7 cm/sec.

ation of particles as performed by the virtual impactor and by the filters in series.

CONCLUSIONS

This work demonstrates that a miniature dichotomous sampler, consisting of a 12 μm Nuclepore filter as the first stage followed by a 0.2 μm Nuclepore filter as the second stage, can separately collect fine and coarse particles from the atmosphere. By choosing an appropriate flow rate, the collection efficiency curve for unit density particles can closely match the ACGIH criteria, but the efficiency curves may deviate for particles of differing specific gravity. For atmospheric sampling, this effect may not be of major significance because the greatest percentage variations in the collection efficiencies occur near the 2 μm minimum of the bimodal distribution of particles. The variation in the collection efficiency curve with specific gravity could be minimized by developing filters with larger pore diameters and operating then at higher face velocity.

Measurements made in this work indicate that a 90 $\mu\text{g}/\text{cm}^2$ loading of atmospheric particles does not affect the filter efficiency near the 50% cutpoint. Further tests are needed at higher loadings.

The series filtration sampler is a promising addition to aerosol sampling technology. However, extensive field testing is needed to delineate fully the capabilities and any limitation of this technique.

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